

# Fabrication and characterization of bismuth–telluride-based alloy thin film thermoelectric generators by flash evaporation method

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## Abstract

Bismuth–telluride-based alloy thin film thermoelectric generators are fabricated by a flash evaporation method. We prepare  $\text{Bi}_{0.4}\text{Te}_{3.0}\text{Sb}_{1.6}$  (p-type) and  $\text{Bi}_{2.0}\text{Te}_{2.7}\text{Se}_{0.3}$  (n-type) powders for the fabrication of the flash evaporated thin films. The overall size of the thin film thermoelectric generators, which consist of seven pairs of legs connected by aluminum electrodes, is 20 mm by 15 mm. Each leg is 15 mm long, 1 mm wide and 1  $\mu\text{m}$  thick. We measure the output voltage and estimate the maximum output power near room temperature as a function of the temperature difference between hot and cold junctions of the thin film thermoelectric generators. In order to improve the performance of the generators, a hydrogen annealing process is carried out at several temperatures from 25 °C to 250 °C. The highest output voltage of 83.3 mV and estimated output power of 0.21  $\mu\text{W}$  are obtained from a hydrogen annealing temperature of  $T_a = 250^\circ\text{C}$  and a temperature difference of  $\Delta T = 30\text{ K}$ . The hydrogen annealing temperature of  $T_a = 250^\circ\text{C}$  also results in the best electrical performance for both p-type thin film (Seebeck coefficient = 254.4  $\mu\text{V/K}$ , resistivity = 4.1  $\text{m}\Omega\text{ cm}$ , power factor = 15.9  $\mu\text{W/cm}^2$ ) and n-type thin film ( $-179.3\text{ }\mu\text{V/K}$ , 1.5  $\text{m}\Omega\text{ cm}$ , 21.5  $\mu\text{W/cm}^2$ ).

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**Keywords:** Thin film; Thermoelectric; Generator; Seebeck coefficient; Power factor; Bismuth–telluride

## 1. Introduction

Recently there have been several applications calling for a few microwatts of power at relatively high-voltage for the operation of small electric devices and systems in the microelectronics industry [1,2]. Even though these electric devices and systems need only a very small amount of power, this has usually been supplied by batteries or external power sources. Under this circumstance, a thermoelectric generator may be an ideal onboard power source. It can directly produce a small electrical power from thermal energy. To apply thermoelectric generators as an alternative power source for small electric devices and systems, the requirements are miniaturization and high performance at room temperature.

To obtain high performance thermoelectric generators, bismuth–telluride-based alloys are widely employed. They are

known to achieve excellent performance at room temperature compared to other thermoelectric materials, and have been used in thermoelectric refrigerators for the temperature control of semiconductor devices such as laser diodes or CCDs (charge coupled devices). The performance of thermoelectric materials depends on the thermoelectric figure-of-merit, ZT, which is defined as  $ZT = S^2 T / \rho \kappa$ , where  $S$  is the Seebeck coefficient,  $\rho$  the electrical resistivity,  $\kappa$  the thermal conductivity, and  $T$  is the absolute temperature. The product  $S^2 / \rho$  is defined as the thermoelectric power factor. The power factor should be maximized and the thermal conductivity should be minimized in order to achieve high efficiency thermoelectric materials.

The primary approach to miniaturize thermoelectric generators is to employ thin film technology. Traditional thermoelectric devices are fabricated from sintered blocks of the materials. However, there are certain difficulties and limitations in making highly miniaturized devices due to the cutting and assembling processes. Thin film technology has a number of deposition methods such as flash evaporation [3–5], co-sputtering [6,7], pulsed laser deposition [8,9], metal organic chemical vapor

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deposition (MOCVD) [10,11] and molecular beam epitaxy (MBE) [12,13]. Here, we employ the flash evaporation method to fabricate bismuth–telluride-based alloy thin film thermoelectric generators. This deposition method has the potential for low production cost than any other deposition methods because of the simple system that consists of only a vacuum chamber with particle holder, tungsten heater, and substrate holder. On the other hand, the performance of the thin films is relatively poor without the annealing process. In fact, annealing processes, especially hydrogen annealing, are known to enhance the transport properties of thermoelectric materials [14,15]. There have been some prior reports of thin film thermoelectric generators [16–18], focusing on attempts to fabricate unique geometric device structures.

In this study, we focus on the effect of annealing conditions to improve the performance of the thin film thermoelectric generators fabricated by a flash evaporation method. We prepare  $\text{Bi}_{0.4}\text{Te}_{3.0}\text{Sb}_{1.6}$  as p-type powders and  $\text{Bi}_{2.0}\text{Te}_{2.7}\text{Se}_{0.3}$  as n-type powders for the fabrication of the flash evaporated thin films. The annealing process is carried out in hydrogen ambient at several temperatures. First, we investigate the intrinsic properties of each of the constituent thin films, and then we measure the output voltage and estimate the maximum output power of a complete generator near room temperature as functions of the temperature difference between hot and cold junctions.

## 2. Experiment

Bismuth–telluride-based alloy thin film thermoelectric generators are fabricated by a flash evaporation method. The flash evaporation equipment is described elsewhere [3]. The thin film thermoelectric generators are deposited on glass substrates (Corning 7059) by using patterned shadow masks for the p- and n-type thin films and their junctions. For flash evaporation, we prepare p-type ( $\text{Bi}_{0.4}\text{Te}_{3.0}\text{Sb}_{1.6}$ ) and n-type ( $\text{Bi}_{2.0}\text{Te}_{2.7}\text{Se}_{0.3}$ ) spherical powders of average size 200  $\mu\text{m}$ . The composition of the powders is confirmed by EDX (energy-dispersive x-ray spectroscopy). The distance between the tungsten heater and the substrate is 200 mm. When the chamber is evacuated to  $1.4 \times 10^{-3}$  Pa, we apply a current of 80 A to the tungsten heater until the substrate temperature reaches 200 °C. The thickness of the deposited thin films is 1.0  $\mu\text{m}$ . We expect that the deposited thin films have the approximately same compositions as those of powders (p-type:  $\text{Bi}_{0.4}\text{Te}_{3.0}\text{Sb}_{1.6}$ , n-type:  $\text{Bi}_{2.0}\text{Te}_{2.7}\text{Se}_{0.3}$ ) because flash evaporation methods are known to have good control of the film composition [4,5]. The junctions between the different legs of the thin film thermoelectric generator are sputtered aluminum with a thickness of 2.0  $\mu\text{m}$ . Fig. 1 shows a schematic illustration and a photograph of the thin film thermoelectric generator. The dimension of p- and n-type legs is 15 mm (length)  $\times$  1.0 mm (width)  $\times$  1.0  $\mu\text{m}$  (thickness) and the spacing between legs is 0.6 mm. The overall size of the generator on the substrate is 22 mm long, 24 mm wide, and 1.0 mm thick, consisting of 7 p/n couples.

In order to improve the performance of the thin film thermoelectric generators, a hydrogen annealing process is performed. The annealing temperatures range from 25 °C to 250 °C for

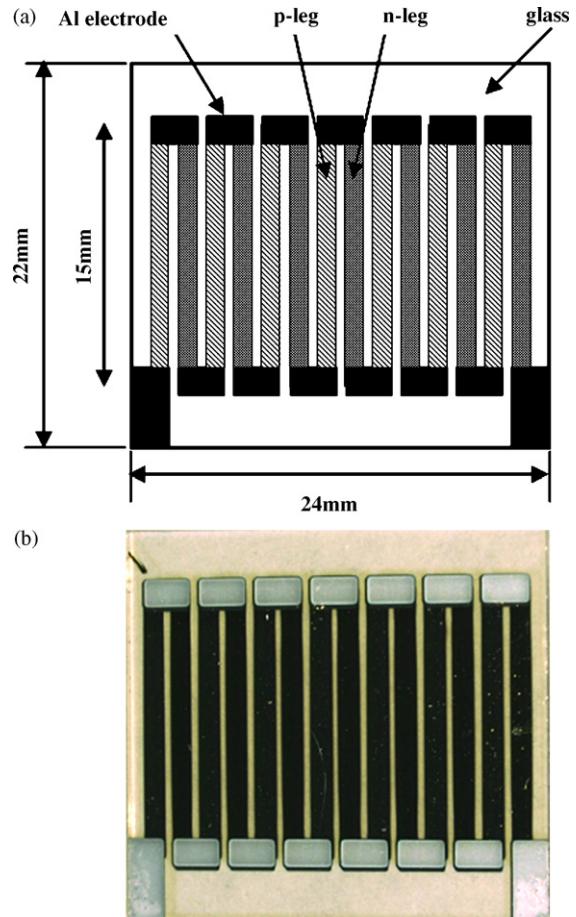


Fig. 1. Schematic illustration (a) and photograph (b) of the bismuth–telluride-based alloy thin film thermoelectric generator.

30 min, in hydrogen ambient at atmospheric pressure. For annealing, samples are placed in an electric furnace that is evacuated to 1.0 Pa and purged five times with high-purity (99.999%) argon gas. Before annealing, the electric furnace is filled with hydrogen gas at atmospheric pressure, and the hydrogen gas flow rate is maintained at 0.3 SLM throughout the annealing process. The temperature is increased steadily at 5 K/min until the temperature reaches the set temperature. After annealing, the samples are cooled down naturally to room temperature. In this study, in order to focus only on the effects of annealing, we use one sample for the entire annealing study. After annealing the sample at 25 °C and measuring the performance of the film, the same sample is annealed at 100 °C and the properties are measured again. This process is repeated for annealing temperatures of 150 °C, 200 °C, and 250 °C. We measure the properties of p- and n-type thin films separately from the overall performance of the generators in order to separate the effects of parasitic resistance. If there is a parasitic resistance in the thin film thermoelectric generators, the output power will be reduced. We first measure the Seebeck coefficient and electrical resistivity of the p- and n-type thin films that make up the generator, and then estimate the thermoelectric power factor expressed as  $\text{P.F.} = S^2/\rho$ , where  $S$  is the Seebeck coefficient and  $\rho$  is the electrical resistivity. The Seebeck coefficient is measured by applying

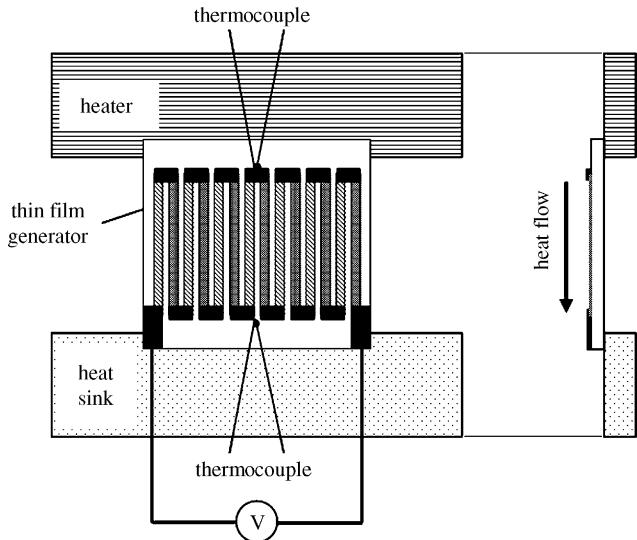


Fig. 2. Schematic diagram of the measurement for the output voltage of the bismuth–telluride-based alloy thin film thermoelectric generators.

a temperature gradient along the in-plane direction while monitoring the resulting Seebeck voltage. The electrical resistivity is measured by four-point probe method. We also investigate the composition of the thin films in our previous study, and may consider that the annealed thin films are essentially stoichiometric [19].

Next we measure the output voltage of the thin film thermoelectric generators while imposing a temperature difference  $\Delta T$  between hot and cold junctions of the generators. The schematic diagram of the measurement for the output voltage is shown in Fig. 2. The both ends of the substrate are contacted with a heater and a heat sink. By applying a current to the heater, the temperature difference is imposed ranging from 5 K to 30 K between hot and cold junctions of the generators. The imposed temperature gradient is parallel to the length of the thermoelectric legs. When we measure the temperatures, thermocouples (chromel–alumel) are attached near the hot and cold junctions near the center of the substrate by pressing the thermocouple bead into an indium ball on the substrate. The thermocouples are not attached directly to the aluminum junctions because this may peel off the aluminum from the substrate when the thermocouples are removed. The output voltage is measured at the aluminum pads connected to the thermoelectric legs. We also measure the overall resistance of the thin film thermoelectric generators by a two-wire method. The maximum output power of the thin film thermoelectric generators is estimated from the output voltage and the overall resistance of the generators. The details are described in the following section.

### 3. Results and discussion

#### 3.1. Transport properties of bismuth–telluride-based alloy thin films

The transport properties of hydrogen annealed thin films, in terms of Seebeck coefficient, the electrical resistivity and the

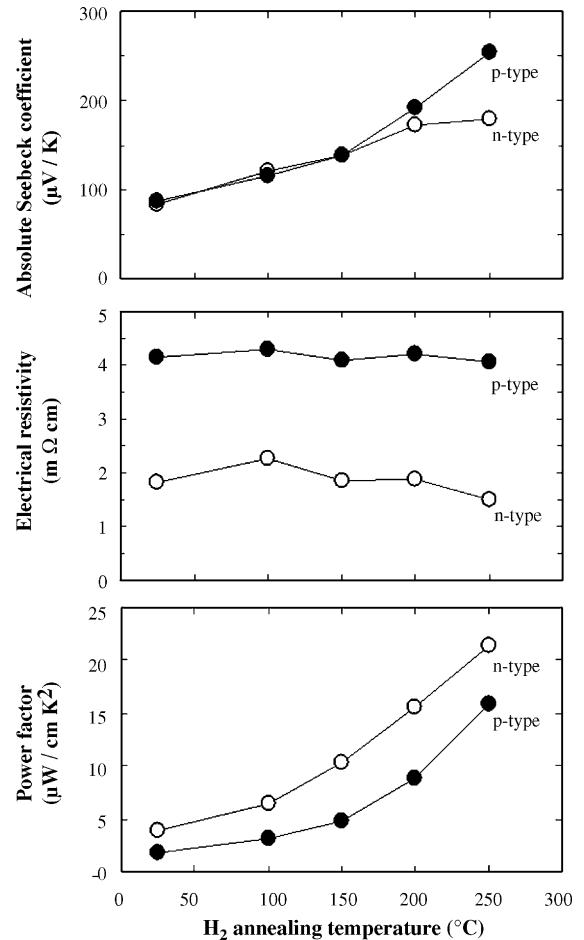


Fig. 3. The transport properties of the bismuth–telluride-based alloy thin film, in terms of Seebeck coefficient, the electrical resistivity, and the thermoelectric power factor, as a function of the hydrogen annealing temperature.

thermoelectric power factor, are investigated at room temperature (Fig. 3). For annealing temperatures of 25 °C to 150 °C, both types of thin films have nearly the same value of the absolute Seebeck coefficient, and which gradually increases as the annealing temperature increases. For annealing temperatures of 150–250 °C, the absolute Seebeck coefficient of the p-type thin films increases more rapidly than that of the n-type thin films. At an annealing temperature of 250 °C, the Seebeck coefficient of the thin films reaches 254.4  $\mu\text{V/K}$  (p-type) and  $-179.3 \mu\text{V/K}$  (n-type).

The electrical resistivity of both types of thin films is almost constant over this range of annealing temperatures. The electrical resistivity of the p-type thin films varies from 4.0  $\text{m}\Omega\text{ cm}$  to 4.2  $\text{m}\Omega\text{ cm}$ , which is two to three times larger than that of the n-type thin films. The behavior of the electrical resistivity of the thin films as a function of hydrogen annealing temperatures is different from its Seebeck coefficient. The Seebeck coefficient of the thin films depends strongly on the annealing temperature, while the electrical resistivity depends only slightly on it. These phenomena are not clear yet, but some reports suggest that the Seebeck coefficient and the electrical resistivity are influenced by the grain sizes and grain boundaries of the thin films [20,21].

Table 1

Transport properties of bismuth–telluride-based alloys

Sample		$S$ ( $\mu\text{V/K}$ )	$\rho$ ( $\text{m}\Omega\text{ cm}$ )	$S^2/\rho$ ( $\mu\text{W/cm K}^2$ )	Reference
Flash evaporated thin films	p-Bi <sub>0.4</sub> Te <sub>3.0</sub> Sb <sub>1.6</sub> ( $T_a = 250^\circ\text{C}$ )	254.4	4.1	15.9	This work
	n-Bi <sub>2.0</sub> Te <sub>2.7</sub> Se <sub>0.3</sub> ( $T_a = 250^\circ\text{C}$ )	-179.3	1.5	21.5	
Co-sputtered thin films	p-BiTeSb	180–190	1.3–1.4	253	[6]
	n-BiTe	-160 to -170	1.6–1.8	15.7	
Single crystals	p-Bi <sub>0.4</sub> Te <sub>3.0</sub> Sb <sub>1.6</sub>	194 ( $S_{11}$ )	1.0 ( $\rho_{11}$ )	37.8	[22]
	n-Bi <sub>2</sub> Te <sub>2.85</sub> Se <sub>0.15</sub>	223 ( $S_{11}$ )	1.1 ( $\rho_{11}$ )	44.8	

The thermoelectric power factor of both p-type and n-type thin films systematically increases as annealing temperatures increase, reaching  $15.9\text{ }\mu\text{W cm}^{-1}\text{ K}^{-2}$  (p-type) and  $21.5\text{ }\mu\text{W cm}^{-1}\text{ K}^{-2}$  (n-type) at a hydrogen annealing temperature of  $250^\circ\text{C}$ . The transport properties of these flash evaporated thin films ( $T_a = 250^\circ\text{C}$ ) are listed in Table 1. We also include the transport properties of co-sputtered thin films and single crystals as references. The subscript (11) of the transport properties of the single crystals stands for the measurement direction that is parallel to the cleavage planes, and these materials achieve higher performance in this direction. Compared to the thermoelectric power factor of the co-sputtered thin films [6], the p-type flash evaporated thin film is inferior but the n-type flash evaporated thin film has an advantage. Therefore, the performance of the flash evaporated thin films in this study is comparable to that of the well-established co-sputtered thin films, so that our results may be considered high performance in the field of thermoelectric thin films. However, the thermoelectric power factors of the flash evaporated thin films are less than half of the values of the single crystals [22]. The deposition conditions of the flash evaporation method should be further optimized to improve the thin film performance.

### 3.2. Characterization of thin film thermoelectric generators

The performance of bismuth–telluride-based alloy thin film thermoelectric generators is investigated near room temperature. By making a temperature difference  $\Delta T$  between hot and cold junctions of the thin film thermoelectric generators, we measure the output voltage and estimate the maximum output power. Fig. 4 shows the output voltage of thin film thermoelectric generators measured as functions of the temperature difference (a) and the hydrogen annealing temperatures (b). The measurement setup is depicted in Fig. 2. The output voltage of all the annealed samples is proportional to the temperature differences over this range of temperatures. It is clear that the output voltage of the thin film thermoelectric generators improves as the hydrogen annealing temperature increases. Thus, we confirm the improvement of the generator performance by the hydrogen annealing process. At the annealing temperature of  $250^\circ\text{C}$ , the output voltage reaches  $83.3\text{ mV}$  at the temperature difference of  $30\text{ K}$ . In this study, the measured output voltages are approximately 10% lower than expected voltages calculated from the Seebeck coefficients of the thin films. This may indicate that we overestimate the true temperature difference, because the measurement is adjacent to both ends of the junctions. We also try

to treat the hydrogen annealing over  $250^\circ\text{C}$ . However, the junctions between the thermoelectric legs and aluminum electrodes break due to thermal stress, and thus the thin film thermoelectric generator does not work. In the future, we need to modify the fabrication and annealing processes of the thin film thermoelectric generators.

The maximum output power of the thin film thermoelectric generators is estimated from the internal resistance and the output voltage of the generators. The internal resistance of the thin film thermoelectric generators is listed in Table 2. The internal resistance consists of the material and parasitic resistance, which includes the resistance of the aluminum electrodes, the electrical contact resistance and the boundary resistance between the aluminum electrodes and the thermoelectric legs. In these gener-

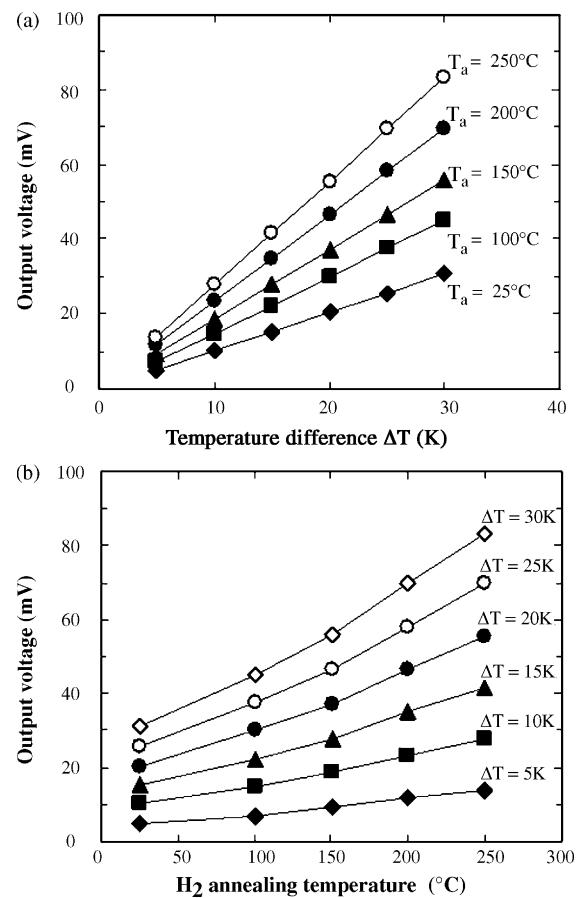


Fig. 4. The generated output voltage of bismuth–telluride-based alloy thin film thermoelectric generators measured as functions of the temperature difference (a) and the hydrogen annealing temperatures (b).

Table 2

Internal resistance of the thin film thermoelectric generators

$T_a$ (°C)	Internal resistance (kΩ)	Material resistance (kΩ)	Parasitic resistance (kΩ)
25	7.6	5.9	1.7
100	7.6	6.4	1.2
150	7.9	5.8	2.1
200	8.3	6.0	2.3
250	8.5	5.5	3.0

ators, the parasitic resistance is mostly attributed to the electrical contact resistance and the boundary resistance because the resistance of aluminum electrodes is expected to be only 0.7–1 Ω, which is less than 0.1% of the whole parasitic resistance. The parasitic resistance of the generators increases as the annealing temperature increases. Generally, the electrical contact resistance reduces as the annealing temperature increases. Therefore, we consider the increasing of the parasitic resistance might be attributed to the boundary resistance between the aluminum electrodes and thermoelectric legs. As we mention previously that the junctions between the thermoelectric legs and aluminum electrodes are broken due to thermal stress at annealing temperature over 250 °C. Even if the annealing temperature is below 250 °C, the micro cracks are expected to form and the resistance increases.

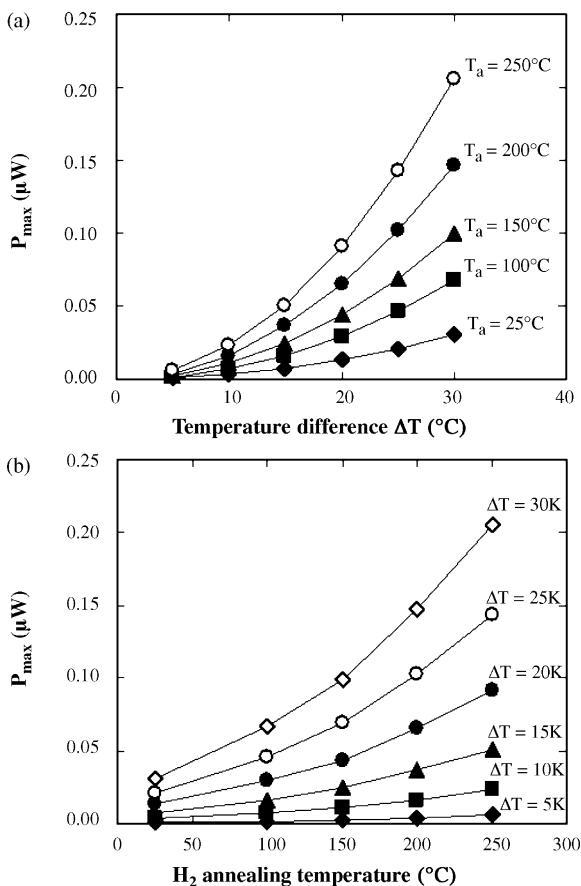


Fig. 5. The estimated maximum output power of bismuth–telluride-based alloy thin film thermoelectric generators measured as functions of the temperature difference (a) and the hydrogen annealing temperatures (b).

The output power can be expressed as  $P = R_L (V_o/R_L + r)^2$ , where  $R_L$  is the external electrical load resistance,  $V_o$  the output voltage, and  $r$  is the internal resistance of the generator including material and parasitic resistance. The material resistance is calculated from their dimensions, the number of thin film couples, and the electrical resistivity. Assuming that the maximum output power is achieved when  $R_L = r$ , the maximum output power can be expressed as  $P_{\max} = V_o^2/4r$ . Fig. 5 shows the maximum output power of thin film thermoelectric generators estimated as functions of the temperature difference (a) and the hydrogen annealing temperatures (b). The maximum output power of all the annealed samples is proportional to the temperature differences in this measured temperature region. It is consistent with the results of the output voltage of the generators. Although the parasitic resistance increases as the hydrogen annealing temperature increases, the maximum output power of the thin film thermoelectric generators is improved. The highest estimated output power of 0.21  $\mu\text{W}$  is obtained for the hydrogen annealing condition of  $T_a = 250^{\circ}\text{C}$  and  $\Delta T = 30\text{ K}$ .

#### 4. Conclusion

Bismuth–telluride-based alloy thin film thermoelectric generators are successfully fabricated by a flash evaporation method, and their performance is improved by a hydrogen annealing process. We measure the performance of the thin film thermoelectric generators near room temperature. The highest output voltage of 83.3 mV and estimated output power of 0.21  $\mu\text{W}$  are obtained from a hydrogen annealing temperature of  $T_a = 250^{\circ}\text{C}$  and a temperature difference of  $\Delta T = 30\text{ K}$ . It is found that the best quality thin films for the generator fabrication can be obtained for p-type thin film ( $S = 254.4\text{ }\mu\text{V/K}$ ,  $\rho = 4.1\text{ m}\Omega\text{ cm}$ ,  $S^2/\rho = 15.9\text{ }\mu\text{W/cm K}^2$ ), and for n-type thin film ( $S = -179.3\text{ }\mu\text{V/K}$ ,  $\rho = 1.5\text{ m}\Omega\text{ cm}$ ,  $S^2/\rho = 21.5\text{ }\mu\text{W/cm K}^2$ ) at a hydrogen annealing temperature of  $T_a = 250^{\circ}\text{C}$ . The maximum output power of the thin film thermoelectric generator in this study is still not enough to apply as a power source for microelectronic devices. However, we provide a promising procedure for fabricating thin film thermoelectric generators by making clear the current difficulties such as the high temperature annealing and the contact resistance. In the future, it should be possible to achieve thin film thermoelectric generators with excellent performance by optimizing the fabrication and annealing processes.

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